Classification of the electronic correlation strength in the Fe-pnictides: The case of the parent compound BaFe$_2$As$_2$

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Electronic correlations in the Fe-pnictide BaFe$_2$As$_2$ are explored within LDA+DMFT, the combination of density functional theory with dynamical mean-field theory. While the correlated band structure is substantially renormalized there is only little transfer of spectral weight. The computed **k**-integrated and **k**-resolved spectral functions are in good agreement with photoemission spectroscopy (PES) and angular resolved PES experiments. Making use of a general classification scheme for the strength of electronic correlations we conclude that BaFe$_2$As$_2$ is a moderately correlated system.

The recent discovery of an entirely new class of high-T_c superconductors based on quasi-twodimensional FeAs-layers [1] rather than on CuO-layers has stimulated intense experimental and theoretical activity. In striking similarity with the high-T_c cuprates, the undoped pnictides are not superconducting under ambient pressure, but exhibit an antiferromagnetic commensurate spin-density wave below 150 K [2]. When electrons (or holes) are added to the system via doping antiferromagnetism is suppressed and superconductivity appears. Since it is generally accepted that the strong Coulomb correlations between the Cu 3d electrons are responsible for the anomalous properties of cuprates, it is tempting to suggest that the same is true for the Fe 3d electrons in pnictides.

Investigations of the Coulomb correlation strength in the Fe-pnictides have arrived at different conclusions. Haule, Shim and Kotliar [3] employed the LDA+DMFT approach [4], the combination of density functional theory (DFT) in the local density approximation (LDA) with the many-body dynamical mean-field theory (DMFT) [5], to study LaO$_{1-x}$F$_x$FeAs. Using a value of the local Coulomb interaction $U = 4$ eV obtained by the random phase approximation (RPA) [6] for metallic iron [7] (which is quite similar to that found by constrained LDA, $U = 3$–4 eV [8, 9, 10]), they concluded that LaO$_{1-x}$F$_x$FeAs is a strongly correlated, bad metal which is close to a Mott metal-insulator transition. A similar conclusion was reached by Craco *et al.* [11] for SmO$_{1-x}$F$_x$FeAs. By contrast, using constrained RPA to compute U in LaFeAsO Nakamura, Arita and Imada [12] obtained smaller values of U, in the range $U = 2.2$–3.3 eV. Therefore they concluded that in LaFeAsO and LaFePO electronic correlations are moderately strong. This was affirmed by Anisimov *et al.* [9] who performed LDA+DMFT investigations for LaFeAsO, with U taken from constrained LDA [13, 14], and compared with photoemission spectroscopy (PES) data. Yet another conclusion was reached by Yang *et al.* [15] who investigate the Fe-pnictides by XAS and RIXS and compared with results from cluster diagonalizations, multiplet and DFT calculations. They estimated the Coulomb interaction as $U \leq 2$ eV and inferred that the Fe-pnictides are weakly correlated systems.

These different assessments raise a general question about the classification of the strength of electronic correlations in a particular material. An overall estimate is provided by the ratio of the local Coulomb interaction U and the band width W. For $U/W < 1$ the system is considered weakly correlated, and the results of DFT approximations are sufficient to explain its electronic and magnetic properties. By contrast, if U is comparable with W or larger, the system is moderately or even strongly correlated, and the Coulomb interactions must be treated explicitly in electronic structure calculations. However, the single parameter U/W is only a rough measure of the electronic correlations; furthermore it is not directly accessible by experiment. Here the effective mass renormalization m^*/m of quasiparticles is a more useful parameter since it permits experimental tests. Nevertheless a single number cannot reliably assess the correlation strength in a real material whose properties are determined by various different aspects such as the electronic band structure, degeneracies, filling, and the Coulomb interaction parameters beyond U. A much more expressive quantity is the **k**-resolved single-particle spectral function $A(\mathbf{k}, \omega)$, and its **k**-integrated variant $A(\omega)$, both of which can be measured by angular resolved photoemission spectroscopy (ARPES) and PES, respectively. They allow for an unambiguous classification of the strength of elec-

tronic correlations. Namely, if $A(\mathbf{k}, \omega)$ is well described by DFT approximations it is justified to call the system *weakly* correlated. If the band structure is substantially renormalized by correlation effects as expressed by an enhancement of the quasiparticle effective mass, but $A(\omega)$ does not yet show incoherent Hubbard bands, the system is *moderately* correlated. Only if the correlation induced transfer of spectral weight is so strong that pronounced Hubbard bands and a distinct quasiparticle peak at the Fermi energy appear, can the system be called *strongly* correlated (a good example is the $3d^1$ system SrVO$_3$ [16]). The spectral functions of correlated electron materials can be computed within the LDA+DMFT framework [4], which provides access to material specific single-particle spectra and higher correlation functions.

In this Letter we employ the LDA+DMFT scheme [4] to explore the importance of electronic correlation effects in BaFe$_2$As$_2$, the undoped parent compound of one of the main pnictide high-T_c superconductor families. For this purpose we calculate the spectral functions $A(\omega)$ and $A(\mathbf{k}, \omega)$ and compare our results with the available experimental data for BaFe$_2$As$_2$. In particular, we apply the scheme described above to classify the correlation strength in this important class of Fe-pnictides.

The LDA+DMFT approach proceeds as follows: First, an effective Hamiltonian $\tilde{h}_{\mathbf{k}}$ is constructed using converged LDA results for the system under investigation, then the many-body Hamiltonian is set up, and finally the corresponding self-consistent DMFT equations are solved. By projecting onto Wannier functions [17] we obtain an effective 16-band Hamiltonian which incorporates five Fe d orbitals and three As p orbitals for each of the two Fe and As ions per unit cell. In the present study we construct Wannier states for a single energy window including both p and d bands. Consequently, the eigenvalues of the effective Hamiltonian $\tilde{h}_{\mathbf{k}}$ exactly correspond to the 16 Fe and As bands from LDA. Thereby hybridization effects between As p and Fe d electrons are explicitly taken into account. The LDA calculations were performed with the experimentally determined crystal structure [18] using the Elk full-potential linearized augmented plane-wave (FP-LAPW) code [19]. Parameters controlling the LAPW basis were kept to their default values. The calculated LDA band structure $\epsilon_{LDA}(\mathbf{k})$ is found to be in good agreement with that of Fink *et al.* [20].

To account for the Coulomb interaction already present in LDA the dd-diagonal elements of the effective Hamiltonian $\tilde{h}_{\mathbf{k}}$ are renormalized by a double counting correction [4] $E_{dc} = U(n_{\text{dmft}} - \frac{1}{2})$,

$$h^{dd}_{\mathbf{k},\alpha\beta} = \tilde{h}^{dd}_{\mathbf{k},\alpha\beta} - E_{dc}\delta_{\alpha\beta}. \qquad (1)$$

Here n_{dmft} is the total, self-consistently calculated number of d electrons per Fe site obtained within the LDA+DMFT. This form of E_{dc} yields reliable results for

transition metal compounds, including the superconductor LaFeAsO [9]. The $p - d$ Hamiltonian to be solved by DMFT then has the form

$$\begin{aligned} H = \sum_{\mathbf{k},\sigma} (&h^{dd}_{\mathbf{k},\alpha\beta} d^{\dagger}_{\mathbf{k}\alpha\sigma} d_{\mathbf{k}\beta\sigma} + h^{pp}_{\mathbf{k},\gamma\delta} p^{\dagger}_{\mathbf{k}\gamma\sigma} p_{\mathbf{k}\delta\sigma} + \\ &h^{dp}_{\mathbf{k},\alpha\gamma} d^{\dagger}_{\mathbf{k}\alpha\sigma} p_{\mathbf{k}\gamma\sigma} + h^{pd}_{\mathbf{k},\gamma\alpha} p^{\dagger}_{\mathbf{k}\gamma\sigma} d_{\mathbf{k}\alpha\sigma}) + \\ &\sum_{i,\sigma,\sigma'} U^{\sigma\sigma'}_{\alpha\beta} n^d_{i\alpha\sigma} n^d_{i\beta\sigma'}. \end{aligned} \qquad (2)$$

Here $d_{\mathbf{k}\alpha\sigma}$ and $p_{\mathbf{k}\gamma\sigma}$ are Fourier transforms of $d_{i\alpha\sigma}$ and $p_{i\gamma\sigma}$, which annihilate the d or p electron with orbital and spin indices $\alpha\sigma$ or $\gamma\sigma$ in the ith unit cell, and $n^d_{i\alpha\sigma}$ is the corresponding occupation number operator.

The DMFT self-consistency equations were solved iteratively for imaginary Matsubara frequencies. The auxiliary impurity problem was solved by the Hirsch-Fye quantum Monte Carlo (QMC) method [21]. The elements of $U^{\sigma\sigma'}_{\alpha\beta}$ matrix are parameterized by U and J according to procedure described in [22]. In the following we use the interaction parameters U=3.1 eV and J=0.81 eV obtained in [9, 10]. Calculations were performed in the paramagnetic state at the inverse temperature $\beta = 1/T$=20 eV^{-1} . The imaginary time interval $0 < \tau < \beta$ was divided into 150 slices, and 6×10^6 QMC sweeps were used in a self-consistency loop within the LDA+DMFT scheme. The local self-energy $\Sigma(\omega)$, which is formally an 16×16 matrix with the only nonzero elements on the diagonal of the dd block, was calculated for real energies ω by analytic continuation from imaginary Matsubara frequencies using the Padé approximant [23].

A quantitative measure of the electron correlation strength is also provided by the quasiparticle renormalization factor $Z = (1 - \frac{\partial\Sigma}{\partial\omega}|_{\omega=0})^{-1}$ which leads to an effective mass enhancement $m^*/m = Z^{-1}$. In general, the self-energy is a matrix, leading to different effective masses for different bands. The masses can be obtained from the self-energy on the real axis, $\Sigma(\omega)$. The calculated m^*/m values for every d-orbital are presented in Table 1. The d$_{x^2-y^2}$ orbital has the smallest effective mass renormalization ($m^*/m = 1.83$). The other d-orbitals have a slightly larger value ($m^*/m = 2.05 - 2.07$). The overall effective mass enhancement $m^*/m \approx 2$ agrees well with the ARPES data for BaFe$_2$As$_2$ [24, 25]. It is also in good agreement with experimental results for the pnictide material LaFePO, where de Haas-van Alphen experiments [26] found a mass enhancement between 1.7 and 2.1, and ARPES [27] measured a band renormalization by a factor of 2.2 compared with the LDA.

The orbitally resolved Fe $3d$ and total As $4p$ spectral functions computed within the LDA and LDA+DMFT, respectively, are compared in Fig. 1. The tetragonal crystal field splitting of the Fe-3d states in this material is rather weak (Δ_{cf}=0.25 eV). Within the LDA all five Fe d orbitals form a common band in the energy range $(-2, +2)$ eV relative to the Fermi level (band width

TABLE I: Effective mass renormalization m^*/m of quasiparticles in BaFe$_2$As$_2$ for different orbitals of the d shell.

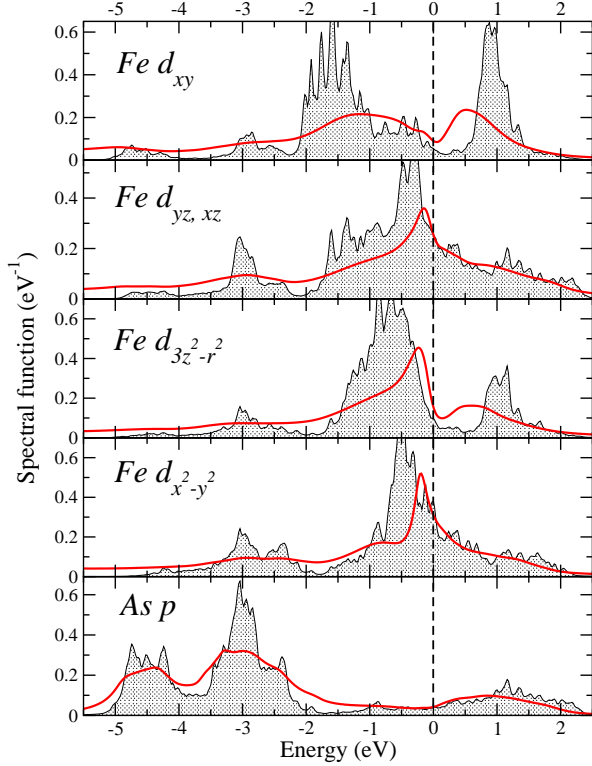| Orbitals	d_{xy}	$d_{yz,xz}$	$d_{3z^2-r^2}$	$d_{x^2-y^2}$
m^*/m	2.06	2.07	2.05	1.83

FIG. 1: (Color online) Orbitally resolved Fe $3d$ and total As p spectral functions of BaFe$_2$As$_2$ obtained within LDA+DMFT (solid lines) are compared with LDA results (shaded areas).

$W \approx 4$ eV). There is a significant hybridization of the Fe t_{2g} orbitals with the As p orbitals, leading to spectral weight in the energy interval $(-3, -2)$ eV where the As p band is located. However, this is not a correlation effect. The correlations only lead to some broadening but not to substantial spectral weight transfer, i.e., there are no Hubbard bands found in the entire spectrum. Indeed, the overall shape of the LDA spectral function is hardly changed by the correlations.

In Fig.2 the total Fe $3d$ and As $4p$ spectral functions are compared with photoemission spectroscopy data of de Jong *et al.* [28]. The computed spectra reproduce the experimental features very well, including the relative positions of the d and p bands and the strong d contribution at the top of the valence band.

We now calculate the **k**-resolved spectral function

$$A(\mathbf{k}, \omega) = -Im\frac{1}{\pi}Tr[(\omega + \mu)\hat{I} - \hat{h}_{\mathbf{k}} - \hat{\Sigma}(\omega)]^{-1}. \quad (3)$$

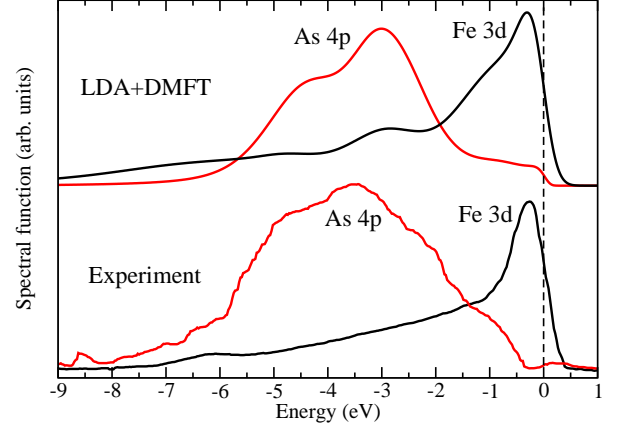Here $\hat{h}_{\mathbf{k}}$ is the 16×16 Hamiltonian matrix on a mesh of

FIG. 2: (Color online) Normalized total Fe d and As p spectral functions of BaFe$_2$As$_2$ calculated within LDA+DMFT (upper curves) are compared with photoemission spectroscopy data of de Jong *et al.* [28] (lower curves).
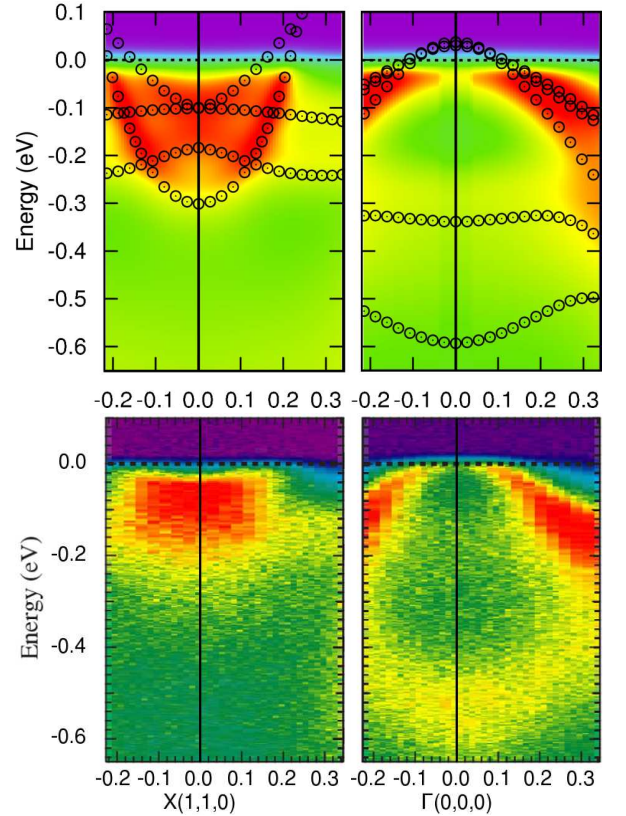

FIG. 3: (Color online) The **k**-resolved total spectral function $A(\mathbf{k}, \omega)$ of BaFe$_2$As$_2$ near the Γ and X points in the Brillouin zone is depicted as a contour plot. Upper panel: LDA+DMFT spectral function including the renormalized band structure (circles) obtained by plotting the peak positions of the spectral function $A(\mathbf{k}, \omega)$. Lower panel: The corresponding experimental ARPES intensity map of Liu *et al.* [29].

k-points and μ is the self-consistently determined chemical potential. In Fig. 3 we compare our results with ARPES data of Liu *et al.* [29]. Both theory and experiment show dispersive bands crossing the Fermi level near the Γ and X points. In addition, two weakly pronounced relatively flat bands can be seen in the region from -0.2 to -0.3 eV and from -0.5 to -0.6 eV near the Γ point. The calculated shape and size of the electron and hole pockets centered at the X and Γ points, respectively, are in good agreement with the ARPES data. Also shown in Fig. 3 (upper panel) is the correlated band structure $\epsilon_{DMFT}(\mathbf{k})$. Near the Fermi energy, i.e., in the energy range from -0.25 eV to zero where quasiparticles are well defined (as expressed by a linear behavior of $\mathrm{Re}\Sigma(\omega)$), this dispersion is very well represented by the scaling relation $\epsilon_{DMFT}(\mathbf{k}) = \epsilon_{LDA}(\mathbf{k})/(m^*/m)$, with m^*/m as the computed mass enhancement. This shows that the band structure is substantially renormalized. Nevertheless, since there is no substantial spectral weight transfer in the spectral function, electronic correlations in $BaFe_2As_2$ are only moderately strong.

In conclusion, by employing the many-body dynamical mean-field theory in combination with LDA electronic band structure calculations (LDA+DMFT approach) we calculated the single-particle **k**-integrated and **k**-resolved spectral functions for $BaFe_2As_2$, a parent compound of the superconducting iron pnictide family. The correlated band structure obtained within LDA+DMFT is well described by the relation $\epsilon_{DMFT}(\mathbf{k}) = \epsilon_{LDA}(\mathbf{k})/(m^*/m)$, with an effective mass enhancement computed as $m^*/m \approx 2$. At the same time correlations do not lead to substantial transfer of spectral weight in the spectrum, i.e., Hubbard bands do not form. Our results are in good agreement with PES experiments of de Jong *et al.* [28] and ARPES data of Liu *et al.* [29]. We also discussed a general classification scheme for the electronic correlation strength based on the renormalization of the band structure and the transfer of spectral weight. Our results obtained for $BaFe_2As_2$ and their comparison with experimental data show that this material, and also other Fe-pnictides, should be classified as *moderately correlated.*

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